HANLE EFFECT FOR MONOCHROMATIC EXCITATION.
NON PERTURBATIVE CALCULATION FOR A J = 0 TO J = 1 TRANSITION

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(Reçu le 3 janvier 1975, accepté le 20 janvier 1975)

Résumé. — On établit des expressions théoriques exactes pour les signaux de fluorescence observés
dans une expérience de croisement de niveaux effectuée avec une excitation monochromatique
sur une transition Jg = 0 → Je = 1. On discute l'effet de l'élargissement radiatif et du désaccord
Zeeman.

Abstract. — Exact theoretical expressions are derived for the fluorescent light observed in a level
crossing experiment performed on a Jg = 0 to Je = 1 transition using monochromatic excitation.
The effect of radiative broadening and Zeeman detuning is discussed.

1. Introduction. — The development of narrow band tunable laser sources results in the possibility
of achieving an almost monochromatic excitation of free atoms. Recently, Rasmussen, Schieder and
Walther observed [1] zero field level crossing resonances on an atomic beam of Ba\(^{138}\) irradiated per-
pendicularly by a single mode laser beam \(\lambda\). The available laser intensity was sufficient to saturate
the atomic transition \(6s^2 \, ^1S_0 \leftrightarrow 6s \, ^1P_1\); in such a case, the radiative broadening of the resonances
may become important and the lowest order theory (with respect to the electric field of the light wave)
is insufficient. It is necessary to go beyond the Born approximation \([2, 3, 4]\) for the scattering amplitude.

The necessity of computing such higher order effects also occurs for a broad-line excitation (spectral
width of the incident beam very large compared to the natural and Doppler widths). In this case, the
shortness of the correlation time of the excitation simplifies the problem : it is possible to obtain rate
equations coupling the density matrices \(\sigma_e\) and \(\sigma_g\) representing the collection of atoms in the excited
and ground states. Their steady state solution gives non perturbative expressions for the various detection
signals and allows a quantitative interpretation of several higher order effects (radiative broadening,
saturation resonances... \([5\) to \(9]\).

For monochromatic excitation, the problem is more difficult : the coherence time of the light is very
long, so that it is impossible to obtain rate equations coupling only \(\sigma_e\) and \(\sigma_g\). The optical coherences \(\sigma_{eg}\)
(off diagonal elements of the density matrix between e and g, which represent the motion of the atomic
dipole moments driven by the incident light wave) must be taken into account, as they are now not
negligible \([9]\). In this case it is better to start from the equations describing the coupled evolution of \(\sigma_e,\)
\(\sigma_g\) and \(\sigma_{eg}\) under the influence of the various processes : Larmor precession, spontaneous emission, interaction
with the laser light, and to try to solve them for arbitrary values of the light intensity. This is what
we do in the present letter, for the simple case of a \(J_g = 0 \rightarrow J_e = 1\) transition (corresponding to the
experiment of reference \([1]\)). From the steady state solution of the equations of evolution, we derive
exact theoretical expressions for various detection signals allowing a quantitative interpretation of
the experimental results.

2. Notations. — We consider an atomic beam propagating along \(Oz\). A magnetic field \(B_0\) is applied
in the same direction. The atoms are irradiated by a single mode laser beam of pulsation \(\omega\) propagating
along \(Oy\) and having a linear polarization \(e_x\) parallel to \(Ox\). One monitors the fluorescence \(L_F(e_y)\) emitted
by the atoms along \(Ox\) with a linear polarization \(e_y\) parallel to \(Oy\). Another possible detection signal
could be the fluorescence \(L_F(e_x)\) emitted along \(Oz\) with a linear polarization parallel to \(Ox\).

The \(J_g = 0\) atomic ground state has only one Zeeman sublevel \(|0\rangle\) which is coupled by the \(\sigma\)-
polarized exciting light to the \(|+1\rangle\) and \(|−1\rangle\)
sublevels of the $J_e = 1$ excited state (we can forget the $m = 0$ excited sublevel). The $|0\rangle \leftrightarrow |+1\rangle$ and $|0\rangle \leftrightarrow |-1\rangle$ transitions correspond to energies respectively equal to $\omega_0 + \Omega_e$ and $\omega_0 - \Omega_e$, where $\omega_0$ is the energy of the optical line in zero magnetic field and $\Omega_e$ the Larmor pulsation in the excited state (we take $\hbar = 1$).

Let $\sigma$ be the density matrix representing the ensemble of atoms in the illuminated zone of the atomic beam. The diagonal elements $\sigma_{++}$, $\sigma_{--}$, $\sigma_{00}$ of $\sigma$ represent the populations of the 3 Zeeman sublevels $|+1\rangle$, $|-1\rangle$, $|0\rangle$. $\sigma_{++} = \sigma_{00}^*$ is the Zeeman coherence between the 2 excited sublevels, $\sigma_{0+} = \sigma_{+0}^*$ and $\sigma_{0-} = \sigma_{-0}^*$ are the optical coherences between $|0\rangle$ and $|+1\rangle$, and $|0\rangle$ and $|-1\rangle$.

From the theory of spontaneous emission, one can show that $L_F(\epsilon_e)$ and $L_F(\epsilon_g)$ are proportional to some linear combinations of the density matrix elements of $\sigma_e$:

$$L_F(\epsilon_e) \propto \sigma_{++} + \sigma_{--} + 2 \text{Re} \sigma_{+-}$$
$$L_F(\epsilon_g) \propto \sigma_{++} + \sigma_{--} - 2 \text{Re} \sigma_{+-}$$

Therefore a quantitative interpretation of the experimental results requires the calculation of $\sigma_{++}$, $\sigma_{--}$, $\sigma_{+-}$.

3. Master equation. — The rate of variation of the various elements of $\sigma$ is given by:

$$\frac{d}{dt} \sigma = -i[H, \sigma]$$

where $H$ is the total Hamiltonian of the system, including the effects of the atomic Hamiltonian $\Omega_0$ (free evolution), and the coupling with the incident beam; some extra terms describing the effect of spontaneous emission must be added to (3). The following equations are easily derived:

$$\dot{\sigma}_{++} = - \Gamma \sigma_{++} - i \Delta \sigma_{0+}$$
$$\dot{\sigma}_{--} = - \Gamma \sigma_{--} + i \Delta \sigma_{0-}$$
$$\dot{\sigma}_{0+} = 2 i \Omega_e \sigma_{-+}$$
$$\dot{\sigma}_{0-} = 2 i \Omega_e \sigma_{+-}$$

where $\Gamma$ is the natural width of the excited state and $\Delta$ is a parameter proportional to the product of the atomic dipole moment by the amplitude $\delta$ of the laser light wave. More precisely,

$$\Gamma = 3 \hbar^2 \delta^2 f_{ge} / 16 \omega_0$$

where $f_{ge}$ is the oscillator strength of the transition $g \rightarrow e$. We have supposed that the laser frequency is not too far from the atomic frequency, so that the so-called rotating wave approximation (valid if $|\omega - \omega_0| \ll \omega_0$ and $\delta \ll \omega_0$) can be used in deriving the theorems of the last column of eq. (4), describing the effect of the coupling between the atoms and the laser. The first column of eq. (4) describes the free evolution of the atoms, the second one, the effect of spontaneous emission (damping of $\sigma_{++}$, $\sigma_{--}$, $\sigma_{+-}$ and $\sigma_{0\pm}$ with a time constant equal to $1/\Gamma$ or $2/\Gamma$, and transfer of atoms from the excited state $e$ to the ground state $g$).

If we put $\sigma_{0\pm} = \rho_{0\pm} e^{i\omega t}$ and $\sigma_{\pm0} = \rho_{\pm0} e^{-i\omega t}$, we can easily transform (4) into a set of linear differential equations with time independent coefficients giving $\dot{\sigma}_{++}$, $\dot{\sigma}_{--}$, $\dot{\sigma}_{0+}$, $\dot{\sigma}_{0-}$ (equivalent of the transformation to the rotating frame in NMR). By equating these rates of variation to zero, one then finds the steady state solution for the various elements of $\sigma$. Reinserting these steady values of $\sigma_{++}$, $\sigma_{--}$, $\sigma_{+-}$, $\sigma_{0\pm}$ into eq. (1) and (2), one finally obtains

the following theoretical expressions for the detection signals $L_F(\epsilon_e)$ and $L_F(\epsilon_g)$:

$$L_F(\epsilon_e) \propto \frac{\Omega_e^2 (D - 1) - \Omega_e Y}{A + (3 D - 1) B}$$
$$L_F(\epsilon_g) \propto \frac{\Omega_e (Y + \Omega_e D) + (D - 1) (\Omega_e^2 + B)}{A + (3 D - 1) B}$$

where $\Delta \omega = \omega - \omega_0$

$$x^2(\Omega_e) = [\langle \gamma \rangle^2 / 2 + (\Delta \omega \pm \Omega_e)^2] \delta^2$$
$$D(\Omega_e) = 1 + (1 + \chi_x)^{-1} + (1 + x^{-})^{-1}$$
$$Y(\Omega_e) = (\Delta \omega - \Omega_e) (1 + \chi_x)^{-1}$$
$$A(\Omega_e) = 4 \Omega_e (Y + \Omega_e D) + 3 Y^2$$
$$B(\Omega_e) = \frac{\Gamma^2}{4} \left( \frac{2}{1 + \chi_x} + \frac{1}{\chi_x} \right) + \left( \frac{\Delta \omega - \Omega_e}{\chi_x} \right)^2 + \left( \Delta \omega + \Omega_e \right)^2 \chi(1 + \chi_x) + \chi^2 (1 + x^2).$$

4. Discussion. — Expressions (5) and (6) give the dependence of the signal on 3 important parameters, the static magnetic field ($\Omega_e$), the light intensity ($\delta^2$), and detuning of the laser from the atomic frequency ($\omega - \omega_0$). If one fixes $\delta^2$ and $\omega - \omega_0$ and varies $\Omega_e$, one obtains the shape of the zero field level crossing resonance. This shape is clearly not lorentzian (for
a broad line excitation, this shape would be always lorentzian in the case of a $J_g = 0 \leftrightarrow J_e = 1$ transition [5, 6, 9]). Figure 1 gives the shape of the level crossing resonances for a zero detuning ($\omega = \omega_0$); each curve corresponds to a different value of the light intensity. One clearly sees a radiative broadening of the resonances. Figure 2 gives the shape of the resonances for a fixed value of the intensity and increasing values of the detuning $\omega - \omega_0$.

\[ L_{\Phi}(e_i) \propto \frac{(\Delta \omega)^2}{(\Omega_e/v)^2 + (\Omega_e/v^2)^2 + 4}. \]  

In this case the shape of the level crossing resonance does not change when $v$ increases, provided that the scale of the horizontal axis is contracted proportionally to $v$. In other words, the height of the resonance saturates and its width increases as the square root of the light intensity. We find a behaviour similar to the one observed with a broad line excitation [5, 6, 9].

When the detuning of the laser is very high ($|\omega - \omega_0| \gg \Gamma, v$), the Zeeman splitting required to put an excited sublevel in resonance with the laser light is so large that Zeeman coherence can no longer appear between the two excited sublevels. Thus $L_{\Phi}(e_i)$ tends to a sum of two Lorentzians centered on $Q_e = \pm (\gamma \omega_0)$, corresponding to the resonant excitation of $|+1\rangle$ or $|-1\rangle$ sublevels:

\[ L_{\Phi}(e_i)_{Q_e} \propto \frac{v^2}{(\Delta \omega - \Omega_e)^2 + 2 v^2 + \Gamma^2/4} \]

\[ + \frac{v^2}{(\Delta \omega + \Omega_e)^2 + 2 v^2 + \Gamma^2/4}. \]

At very high magnetic field

\[ (Q_e \gg \Gamma, |\omega - \omega_0|, v) \quad L_{\Phi}(e_i) \] and $L_{\Phi}(e_i)$

tend to zero. This is due to the fact that the frequencies $\omega_\pm \pm Q_e$ of the optical lines $0 \leftrightarrow +1$ and $0 \leftrightarrow -1$ are out of resonance with the laser frequency $\omega$. Let us recall that for a broad line excitation, $L_{\Phi}$ would tend to a non-zero limiting value corresponding to the light reemitted incoherently from $|+1\rangle$ and $|-1\rangle$ sublevels.

The preceding calculation could be easily generalized to atomic transitions corresponding to higher values of $J_g$ and $J_e$. It seems difficult to get explicit analytical expressions for the steady state solution of the coupled equations of evolution of $\sigma_{\Phi}, \sigma_{\Phi}', \sigma_{\Phi''}$, but, as these equations are linear, it is always possible to compute this solution, at least numerically, by inverting a matrix.

Acknowledgments. — The authors would like to thank Professor H. Walther for communicating the experimental results of his group before publication.
References


